



IN THE UNITED STATES PATENT AND TRADE MARK OFFICE

In re Application of OKI, Jun et al.

Serial No. 10/568,779

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Group Art Unit : 1621

Examiner : CARR, DEBORAH D

ESTERIFICATION PRODUCT AND COSMETICS

\* \* \* \* \*

DECLARATION

Honorable Commissioner of  
Patent and Trademarks  
Washington, D.C. 20231

I, Yasunori NOGUCHI, a citizen of Japan, residing at 2-2-16,  
Yasaka-cho, Kishiwada-shi, Osaka, Japan, declare and say:

that I graduated from Department of Applied Chemistry,  
Faculty of Engineering, Osaka Institute of Technology, in March,  
1987 and since October, 1989, I have been employed by Sakamoto  
Yakuhin Kogyo Co., Ltd., where I have been engaged in research  
and development of Chemicals for Cosmetics in the Research  
Laboratories of said Company;

that I am one of the co-inventors of the above-identified  
application

that I am familiar with the invention and the prosecution  
history of the above-identified application; and

that in order to demonstrate the superiority of the  
esterification products of the invention claimed in the  
above-identified application, I beg to submit the following  
experimental data which have been obtained under my supervision:

## Experiment

### 1. Production of a polyglycerin ester of fatty acid

#### SAMPLE 1 (Same as EXAMPLE 1 in the specification)

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 337 g of isostearic acid were charged. Following addition of 0.2 g of sodium hydroxide, an esterification reaction was carried out at 250°C for 4 hours under nitrogen flow. Thus, 395 g of esterification product of branched fatty acid was produced. The esterification product of branched fatty acid had a degree of esterification of 75.0% and an acid value of 1.0. This esterification product is named as SAMPLE 1.

#### SAMPLE 2

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 337 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1, except for changing the reaction time(\*). Thus, 395 g of esterification product of branched fatty acid was produced. The esterification product of branched fatty acid had a degree of esterification of 75.0% and an acid value of 3.0. This esterification product is named as SAMPLE 2.

\* In the Productions of SAMPLES 1-18, the reaction times were not the same each other, even if being mentioned, "a reaction was carried out under conditions the same as . . . . .". Some of the reaction times were changed for changing the acid value.

#### SAMPLE 3

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value

and 427 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 475 g of esterification product of branched fatty acid was produced. The esterification product of branched fatty acid had a degree of esterification of 95.0% and an acid value of 3.0. This esterification product is named as SAMPLE 3.

**SAMPLE 4 (Same as EXAMPLE 2 in the specification)**

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 6 calculated on the basis of a hydroxyl value and 150 g of 2-ethylhexanoic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 220 g of esterification product of branched fatty acid was produced. The esterification product of branched fatty acid had a degree of esterification of 60.0% and an acid value of 0.5. This esterification product is named as SAMPLE 4.

**SAMPLE 5**

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 6 calculated on the basis of a hydroxyl value and 150 g of caprylic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 219 g of esterification product of branched fatty acid was produced. The esterification product of branched fatty acid had a degree of esterification of 60.0% and an acid value of 1.0. This esterification product is named as SAMPLE 5.

**SAMPLE 6 (Same as EXAMPLE 3 in the specification)**

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 12 calculated on the basis of a hydroxyl value and 351 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 407 g of esterification product of branched fatty acid was produced. The esterification product of branched fatty acid had a degree

of esterification of 80.0% and an acid value of 3.0. This esterification product is named as SAMPLE 6.

SAMPLE 7 (Same as COMPARATIVE EXAMPLE 1 in the specification)

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 225 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 295 g of esterification product of branched fatty acid was produced. The esterification product of branched fatty acid had a degree of esterification of 50.0% and an acid value of 5.0. This esterification product is named as SAMPLE 7.

SAMPLE 8 (Same as COMPARATIVE EXAMPLE 2 in the specification)

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 4 calculated on the basis of a hydroxyl value and 380 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 433 g of esterification product of branched fatty acid was produced. The esterification product of fatty acid had a degree of esterification of 70.0% and an acid value of 1.0. This esterification product is named as SAMPLE 8.

SAMPLE 9

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 337 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 393 g of esterification product of branched fatty acid was produced. The esterification product of fatty acid had a degree of esterification of 75.0% and an acid value of 5.0. This esterification product is named as SAMPLE 9.

SAMPLE 10 (Same as COMPARATIVE EXAMPLE 3 in the specification)

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 335 g of oleic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 393 g of esterification product was produced. The esterification product of fatty acid had a degree of esterification of 75.0% and an acid value of 1.0. This esterification product is named as SAMPLE 10.

SAMPLE 11 (Same as COMPARATIVE EXAMPLE 4 in the specification)

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 337 g of stearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 395 g of esterification product was produced. The esterification product of fatty acid had a degree of esterification of 75.0% and an acid value of 1.0. This esterification product is named as SAMPLE 11.

SAMPLE 12

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 292 g of stearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 355 g of esterification product was produced. The esterification product of fatty acid had a degree of esterification of 65.0% and an acid value of 1.0. This esterification product is named as SAMPLE 12.

SAMPLE 13

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 270 g of stearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 335

g of esterification product was produced. The esterification product of fatty acid had a degree of esterification of 60.0% and an acid value of 3.0. This esterification product is named as SAMPLE 13.

#### SAMPLE 14

Lanolin commercially available is used as SAMPLE 14.

#### SAMPLE 15

Diglyceryl triisostearate commercially available is used as SAMPLE 15.

#### SAMPLE 16

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 270 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 335 g of esterification product was produced. The esterification product of fatty acid had a degree of esterification of 60.0% and an acid value of 3.0. This esterification product is named as SAMPLE 16.

#### SAMPLE 17

Into a reactor, 100 g of polyglycerol having an average degree of polymerization of 10 calculated on the basis of a hydroxyl value and 270 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 335 g of esterification product was produced. The esterification product of fatty acid had a degree of esterification of 60.0% and an acid value of 4.0. This esterification product is named as SAMPLE 17.

#### SAMPLE 18

Into a reactor, 100 g of polyglycerol having an average degree

of polymerization of 10 calculated on the basis of a hydroxyl value and 225 g of isostearic acid were charged. Then, a reaction was carried out under conditions the same as those for SAMPLE 1. Thus, 295 g of esterification product was produced. The esterification product of fatty acid had a degree of esterification of 50.0% and an acid value of 3.0. This esterification product is named as SAMPLE 18.

## 2. Evaluation of Water-holding property and Odor

Evaluation of water-holding property and odor of each of SAMPLES 1-18 were carried out according to a test of percentage of water hold and a test of odor intensity, each being provided below.

### [Test of Percentage of Water Hold]

A mixture of 1 g of each of SAMPLES 1-18 and 9 g of vaseline was used as a test sample. While the test sample was stirred, water was added thereto dropwise slowly. Thus, the test sample was caused to hold water until water started oozing. The percentage of water hold of this case was defined by:  $\text{percentage of water hold (\%)} = \frac{\text{weight of water hold (g)}}{\text{weight of test sample (g)}} \times 100$ .

### [Test of Odor Intensity]

On the back of a hand, 1 g of esterification product of each of SAMPLES 1-18 was spread and then the odor intensity was evaluated sensorily according to the criteria given below. The number of the monitors who carried out the sensory evaluation was twenty including men and women.

- Criteria
- 5 : No odor is perceived.
  - 4 : Almost no odor is perceived.
  - 3 : Weak odor is perceived.
  - 2 : Odor is perceived.
  - 1 : Strong odor is perceived.

The results of the test of percentage of water hold (in the Table, indicated as "Percentage of water hold") and the results of the test of odor intensity (average among the monitors. in the Table, indicated as "Odor") are shown in the Table together with the average degree of polymerizations of the polyglycerols used in the esterifications, the fatty acids, the degrees and acid values of esterification of the esterification products.

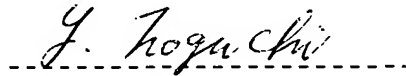


**Table**

Sample No	Average degree of polymerization of polyglycerol	Fatty acid	Degree of esterification (%)	Acid value	Percentage of water hold (%)	Odor
1	1 0	Isostearic acid	75. 0	1. 0	560	4. 9
2	1 0	Isostearic acid	75. 0	3. 0	550	4. 0
3	1 0	Isostearic acid	95. 0	3. 0	280	3. 9
4	6	2-ethyl-hexanoic acid	60. 0	0. 5	510	4. 6
5	6	Caprylic acid	60. 0	0. 5	150	1. 2
6	1 2	Isostearic acid	80. 0	3. 0	580	4. 0
7	1 0	Isostearic acid	50. 0	5. 0	330	1. 6
8	4	Isostearic acid	70. 0	1. 0	200	4. 8
9	1 0	Isostearic acid	75. 0	5. 0	510	1. 5
10	1 0	Oleic acid	75. 0	1. 0	300	2. 7
11	1 0	stearic acid	75. 0	1. 0	180	4. 8
12	1 0	stearic acid	65. 0	1. 0	170	4. 9
13	1 0	stearic acid	60. 0	3. 0	150	4. 0
14 Lanolin	—	—	—	—	360	1. 0
15	2	Isostearic acid	75. 0	1. 0	290	4. 7
16	1 0	Isostearic acid	60. 0	3. 0	400	4. 1
17	1 0	Isostearic acid	60. 0	4. 0	380	3. 2
18	1 0	Isostearic acid	50. 0	3. 0	330	4. 0

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United State Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 26<sup>th</sup> day of November 2007.

A handwritten signature in cursive script, appearing to read 'Y. Noguchi', is written over a horizontal dashed line.

Yasunori NOGUCHI